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# Nonclassical photon statistics in spontaneous Raman and in stimulated Raman processes

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**Abstract** : The equations of motion of the field operators corresponding to the spontaneous and stimulated Raman processes give rise to the coupled nonlinear differential equations. The solutions of these coupled differential equations involving field operators are used to calculate the second order correlation function for zero time delay and hence the photon-bunching, photon-antibunching and nonclassical photon statistics of the input coherent light.

**Keywords** : Stimulated Raman scattering, photon antibunchings, sub-poissonian, photon statistics

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## 1. Introduction

In their experiment, Hanbury-Brown and Twiss [1] obtained the intensity correlation of an incandescent light source and they concluded that the photons come together. The phenomenon in which the photons come together is known as photon bunching. With the availability of new radiation sources, the people are getting more and more involved in the studies of the quantum statistical properties (QSP) of the radiation field. In addition to the usual photon-bunching and photon antibunching, the QSP provide more information about the detailed nature of the radiation field. For example, the photon number distribution could be obtained from the knowledge of QSP of the radiation field. In order to study the QSP, we calculate the second order correlation function for zero time delay [2–4]

$$g^2(0) = \frac{< a^\dagger a^\dagger aa >}{< a^\dagger a >^2}, \quad (1)$$

where $a$ ($a^\dagger$) is the usual annihilation (creation) operator of the radiation field. Now, for thermal/chaotic light $g^2(0) > 1$ and hence the photons are bunched. Interestingly, the radiation field prepared in coherent state gives rise to $g^2(0) = 1$. It is quite reasonable to guess that there should be some radiation sources with $g^2(0) < 1$ and hence the photon antibunching. The bunching of photons could be explained by using both the wave and particle nature of the light and, therefore, it is regarded as the classical phenomenon. On the other hand, the antibunching of photons could only be explained by using a quantum (photon) nature of light. For this reason, the antibunching of photons is regarded as a purely quantum mechanical phenomenon without classical analogue. On the basis of several experimental observation, it is already established that there is a natural tendency of photons to come together (*i.e.* bunching of photons) [1–3]. In spite of the natural tendency of bunching of photons, there are several predictions where the photons show the anti-bunching effects. For example, various coherent states [4] and the degenerate parametric amplifier [5] were predicted as possible sources of getting antibunched light. In addition to this, the resonance fluorescence from a two level atom [6], the two photon Dick model [7] and the coherent light interacting with a two-photon medium [8] could also be the possible sources for producing antibunched light. The fluorescence spectrum of a single two level atom shows the antibunching and the squeezing as well [6]. There are several examples where the antibunched light are produced in the laboratory [9–20]. In most of these experiments [11–15], the resonance fluorescence from a

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small number of ions [11] atoms or molecules [12–15] are studied to obtain the antibunching effects. The basic physics behind these experiments are clear. The atoms (or molecules) emit radiation and go to a ground state from where no subsequent radiation is possible. Hence the photons are antibunched. The resonance fluorescence field from many-atom source is not suitable for antibunching of photons since the photons emitted are highly uncorrelated. However, a suitable phase matching condition similar to that of four-wave mixing leads to the photon antibunching even in the resonance fluorescence of a multi atomic system [16].

A one more useful parameter in the context of QSP of the radiation field is the *Mandel Q-parameter* and is defined as [2–4]

$$q=\frac{(\Delta N)^2-<N>}{<N>}=<N>(g^2(0)-1), \tag{2}$$

where $<N>$ is the average number of photons present in the radiation field. The usual second order variance in photon number is $(\Delta N)^2 = <N^2> - <N>^2$. We write the numerator $D = (\Delta N)^2 - <N>$ . It is obvious that $Q = 0$, for $g^2(0) = 1$ and the corresponding photon number distribution (PND) is called Poissonian. The Poissonian PND is a characteristic of coherent state [21]. In case of $Q < 0$ and $Q > 0$ the corresponding PNDs are called sub-Poissonian and super-Poissonian respectively. It is normally seen that the photon antibunching comes along with the sub-Poissonian photon statistics. However, there is no such rule that the photon antibunching (bunching) will show the sub-Poissonian (super-Poissonian) photon statistics [22,11].

The quantum statistical properties of the radiation field in stimulated and in spontaneous Raman processes are not investigated properly. Therefore, in the present investigation we will take care the quantum statistical properties of the radiation field in stimulated and in spontaneous Raman processes.

## 2. The model Hamiltonian

The stimulated Raman scattering (SRS) is a direct resonant interaction between incident laser field and the active medium. During the Raman scattering processes, the active medium makes transition to produce vibrational phonon. As a matter of fact, the third order nonlinear medium could be used to obtain the stimulated and spontaneous Raman processes. Now, we consider that a single mode of the vibration phonon is produced. In such a situation, the Hamiltonian corresponding to the spontaneous Raman and stimulated Raman processes are described by the following equation

$$\begin{aligned} H &= \omega_a a^\dagger a+\omega_b b^\dagger b+\omega_c c^\dagger c+\omega_d d^\dagger d \\ &\quad +g(ab^\dagger c^\dagger + h.c.)+\chi(acd^\dagger + h.c), \end{aligned} \tag{3}$$

where *h.c.* stands for the ***Hermitian conjugate***. In the present investigation, we use $h = 1$. The annihilation (creation) operators $a(a^\dagger)$, $b(b^\dagger)$, $c(c^\dagger)$ and $d(d^\dagger)$ correspond the laser (pump) mode, Stokes mode, vibration phonon mode and anti-Stokes mode respectively. Note that the operators $a$, $b$, $c$ and $d$ are commuting in nature. However, $[a, a^\dagger] = [b, b^\dagger] = [c, c^\dagger] = [d, d^\dagger] = 1$. The parameters $\omega_a$, $\omega_b$, $\omega_c$ and $\omega_d$ correspond the frequencies of the modes $a$, $b$, $c$ and $d$ respectively. The first four terms in the right hand side of eq. (3) correspond to the free field parts of the Hamiltonian $H$. The remaining terms under the parentheses correspond to the interaction terms. The parameters $g$ and $\chi$ are the Stokes and anti-Stokes coupling constants respectively. Without loss of generality, we assume that the coupling constants $g$ and $\chi$ are real. We obtain the Heisenberg operator equations of motion involving the field operators

$$\begin{aligned} \dot{a} &= -i(\omega_a a+gbc+\chi c^\dagger d), \\ \dot{b} &= -i(\omega_b b+gac^\dagger), \\ \dot{c} &= -i(\omega_c c+gab^\dagger+\chi a^\dagger d), \\ \dot{d} &= -i(\omega_d d+\chi ac). \end{aligned} \tag{4}$$

The purpose of the present paper is to carried an analytical investigation of photon-bunching, photon-antibunching and the nonclassical photon statistics of the input coherent light. Obviously, the coupled differential equations (4) do not provide exact analytical solutions. Therefore, we adopt an intuitive approach to obtain approximate analytical solutions to the field operators corresponding to eq. (4). As a matter of fact, the solution is already available in our recent work on the squeezing effects of the input coherent light coupled to a nonlinear medium responsible for spontaneous Raman and stimulated Raman processes.

For strong pump (*i.e.* classical pump) field, eqs. (4) may be solved in a closed analytical form [23] and for weak pump field, the value of the nonlinear coupling constant $\chi$ and $g$ are small and the perturbative solution for field operators are possible [24–26]. In these cases, the presence of $\chi$ and $g$ are treated as small perturbation. Hence, it is possible to neglect the higher powers of $\chi$

and $g$ beyond the quadratic one. As a matter of fact, it is envisaged that the short-time approximation is necessary to obtain the analytical solutions to the differential eqs. (4) [24–26]. However, we find that the short-time approximation should be supplemented or need to be modified for the physical situations where the interaction time are quite large. In the present investigation, we use the intuitive approach (IA) for getting the solutions to the differential equations (4). Under weak pump approximation, the solutions of the differential equations (4) assume the following form [27] :

$$
\begin{aligned}
a(t) &= f_1 a(0) + f_2 b(0)c(0) + f_3 c^\dagger(0)d(0) \\
&\quad + f_4 a^\dagger(0)b(0)d(0) + f_5 a(0)b(0)b^\dagger(0) \\
&\quad + f_6 a^\dagger(0)c^\dagger(0)c(0) + f_7 a(0)c^\dagger(0)c(0) \\
&\quad + f_8 a(0)d^\dagger(0)d(0), \\
b(t) &= g_1 b(0) + g_2 a(0)c^\dagger(0) \\
&\quad + g_3 a^2(0)d^\dagger(0) + g_4 c^{\dagger 2}(0)d(0) \\
&\quad + g_5 b(0)c(0)c^\dagger(0) + g_6 b(0)a(0)a^\dagger(0), \\
c(t) &= h_1 c(0) + h_2 a(0)b^\dagger(0) + h_3 a^\dagger(0)d(0) \\
&\quad + h_4 b^\dagger(0)c^\dagger(0)d(0) + h_5 c(0)a(0)a^\dagger(0) \\
&\quad + h_6 c(0)b(0)b^\dagger(0) + h_7 c(0)d^\dagger(0)d(0) \\
&\quad + h_8 c(0)a^\dagger(0)a(0), \\
d(t) &= l_1 d(0) + l_2 a(0)c(0) + l_3 a^2(0)b^\dagger(0) + l_4 b(0)c^2(0) \\
&\quad + l_5 c^\dagger(0)c(0)d(0) + l_6 a(0)a^\dagger(0)d(0). 
\end{aligned} \tag{5}
$$

The corresponding $f_i(t)$ are

$$
\begin{aligned}
f_1 &= \exp(-i\omega_a t), \\
f_2 &= \frac{g e^{-i\omega_a t}}{\Delta\omega_1}\left[e^{-i\Delta\omega_1 t} - 1\right], \\
f_3 &= -\frac{\chi e^{-i\omega_a t}}{\Delta\omega_2}\left[e^{-i\Delta\omega_2 t} - 1\right], \\
&\quad e^{-i\omega_a t} \quad \frac{e^{-i(\Delta\omega_1 - \Delta\omega_2)t} - 1}{\Delta\omega_1 - \Delta\omega_2} + \frac{e^{i\Delta\omega_2 t}}{\Delta\omega_2} \\
&\quad \frac{\chi g e^{-i\omega_a t}}{\Delta\omega_2} \quad \frac{e^{-i(\Delta\omega_1 - \Delta\omega_2)t} - 1}{\Delta\omega_1 - \Delta\omega_2} + \frac{e^{i\Delta\omega_1 t}}{\Delta\omega_1} \\
f_5 &= \frac{g^2 e^{i\omega_a t}}{\Delta\omega_1^2}\left[e^{i\Delta\omega_1 t} - 1\right] - \frac{i g^2 t e^{i\omega_a t}}{\Delta\omega_1} \\
f_6 &= f_5, \\
f_7 &= \frac{\chi^2 e^{-i\omega_a t}}{\Delta\omega_2^2}\left[e^{-i\Delta\omega_2 t} - 1\right] - \frac{i\chi^2 t e^{-i\omega_a t}}{\Delta\omega_2} \\
f_8 &= f_7,
\end{aligned} \tag{6}
$$

where $\Delta\omega_1 = (\omega_b + \omega_c - \omega_a)$ and $\Delta\omega_2 = (\omega_a + \omega_c - \omega_d)$. Clearly, the solution (6) are restricted up to the second orders in $g$ and $\chi$. The parameter $f_1$ is zeroth order in $g$ and $\chi$ and is a free evolving term. The parameters $f_2$ and $f_3$ are first order in $g$ and $\chi$, respectively. The remaining parameters $f_i/(i$ = 4, 5, 6, 7 and 8) are second order in $g$ and/or in $\chi$. The analytical expressions for other coefficients are irrelevant for the present investigation and may be found in our recent publication [27]. It is clear that the detuning $\Delta\omega_1$ and $\Delta\omega_2$ are extremely small. As a matter of fact, the values of $\Delta\omega_1$ and $\Delta\omega_2$ vanish identically if there is no damping in the system. In the present investigation, we however assume that the small damping is present and hence $\Delta\omega_1 \neq 0$ and $\Delta\omega_2 \neq 0$. In the present investigation, we use $|\Delta\omega_1|$ = 0.0001 and $|\Delta\omega_2|$ = 0.00081.

### 3. Photon bunching and photon antibunching

In this section, we would like to investigate the photon bunching, photon antibunching and the nonclassical photon statistics of the input coherent light coupled to the nonlinear medium responsible for spontaneous Raman and stimulated Raman processes. The initial composite coherent state obey the following eigenvalue equations

$$
\begin{aligned}
a(0)\left|\{|\alpha\rangle|\alpha_1\rangle|\alpha_2\rangle|\alpha_3\rangle\}\right\rangle &= \alpha\left|\{|\alpha\rangle|\alpha_1\rangle|\alpha_2\rangle|\alpha_3\rangle\}\right\rangle, \\
b(0)\left|\{|\alpha\rangle|\alpha_1\rangle|\alpha_2\rangle|\alpha_3\rangle\}\right\rangle &= \alpha_1\left|\{|\alpha\rangle|\alpha_1\rangle|\alpha_2\rangle|\alpha_3\rangle\}\right\rangle, \\
c(0)\left|\{|\alpha\rangle|\alpha_1\rangle|\alpha_2\rangle|\alpha_3\rangle\}\right\rangle &= \alpha_2\left|\{|\alpha\rangle|\alpha_1\rangle|\alpha_2\rangle|\alpha_3\rangle\}\right\rangle, \\
d(0)\left|\{|\alpha\rangle|\alpha_1\rangle|\alpha_2\rangle|\alpha_3\rangle\}\right\rangle &= \alpha_3\left|\{|\alpha\rangle|\alpha_1\rangle|\alpha_2\rangle|\alpha_3\rangle\}\right\rangle,
\end{aligned} \tag{7}
$$

where $\alpha$, $\alpha_1$, $\alpha_2$ and $\alpha_3$ are the complex amplitudes corresponding to the pump, Stokes, vibrational phonon and anti-Stokes modes, respectively. Therefore, the number of photons present in the pump, Stokes, vibrational phonon and in anti-Stokes mode are $|\alpha|^2$, $|\alpha_1|^2$, $|\alpha_2|^2$ and $|\alpha_3|^2$ respectively. Now, we give the explicit expression for the number operator for pump mode

$$
\begin{aligned}
N_a = {} & |f_1|^2 a^\dagger(0)a(0) + |f_2|^2 b^\dagger(0)c^\dagger(0)b(0)c(0) \\
& + |f_3|^2 c(0)d(0)^\dagger c^\dagger(0)d(0) + \Big[ f_1^* f_2 a^\dagger(0)b(0)c(0) \\
& + f_1^* f_3 a^\dagger(0)c^\dagger(0)d(0) + f_1^* f_4 a^\dagger(0)a^\dagger(0)b(0)d(0) \\
& + f_1^* f_5 \left( a^\dagger(0)a(0) + a^\dagger(0)a(0)b^\dagger(0)b(0) \right) \\
& + f_1^* f_6 a^\dagger(0)a(0)c^\dagger(0)c(0) \\
& + f_1^* f_7 a^\dagger(0)a(0)c^\dagger(0)c(0) \\
& + f_1^* f_8 a^\dagger(0)a(0)d^\dagger(0)d(0) \\
& + f_2^* f_3 b^\dagger(0)c^{\dagger 2}(0)d(0) + c.c \Big].
\end{aligned}
\tag{8}
$$

Now, we calculate the second order variance of number operator for pump mode $a$

$$
\begin{aligned}
(\Delta N_a)^2 = {} & |f_1|^4 |\alpha|^2 + |f_1|^2 |f_2|^2 \\
& \left( |\alpha|^2 + |\alpha|^2|\alpha_1|^2 + |\alpha|^2|\alpha_2|^2 + |\alpha_1|^2|\alpha_2|^2 \right) \\
& + |f_1|^2|f_3|^2 \left( |\alpha_3|^2 + |\alpha|^2|\alpha_2|^2 + 3|\alpha|^2|\alpha_3|^2 + |\alpha_2|^2|\alpha_3|^2 \right) \\
& + |f_1|^2 \Big( f_1^* f_2 \alpha^* \alpha_1 \alpha_2 + f_1^* f_3 \alpha^* \alpha_2^* \alpha_3 \\
& + 2 f_1^* f_4 \alpha^* \alpha^* \alpha_1 \alpha_3 + 2 f_1^* f_5 \left( |\alpha|^2 + |\alpha|^2|\alpha_1|^2 \right) \\
& + 2 f_1^* f_6 |\alpha|^2 |\alpha_2|^2 + 2 f_1^* f_7 |\alpha|^2 |\alpha_2|^2 \\
& + 2 f_1^* f_8 |\alpha|^2 |\alpha_3|^2 + f_2^* f_3 \alpha_1 \alpha_2^2 \alpha_3^* + c.c. \Big) \\
& \left( f_1^{*2} f_2 f_3 \alpha^{*2} \alpha_1 \alpha_3 + c.c. \right).
\end{aligned}
\tag{9}
$$

In order to investigate the nonclassical photon statistics for pump mode, we calculate the second order correlation function for zero time delay $g_a^2(0)$. The subscript $a$ stands for the pure mode $a$. In a similar manner, it is possible to define the second order correlation functions for other modes $b$, $c$, and $d$. Of course, in the present investigation we keep ourselves confined only for the pump mode. For $g_a^2(0) > 1$, the photons in the filed mode $a$ are said to be bunched. On the other hand the photons are antibunched if $g_a^2(0) < 1$. In other words, the antibunching of photons are possible when $D_a < 0$. It is because the denominator $<N_a>^2$ is always positive. Again the bunching of photons will be observed for $D_a \geq 0$. By using the solutions for the field operators for various modes, we readily obtain the analytical expressions for $D_a$,

$$
\begin{aligned}
D_a = {} & (\Delta N_a)^2 - \langle N_a \rangle = |f_2|^2 |\alpha|^2 \left( 1 + |\alpha_1|^2 + |\alpha_1|^2 \right) \\
& + |f_3|^2 |\alpha|^2 \left( |\alpha_2|^2 + |\alpha_3|^2 \right) \\
& + \left[ \alpha^{*2} \alpha_1 \alpha_3 \left( f_1^* f_4 + f_1^{*2} f_2 f_3 \right) + c.c. \right] \\
& + \Big[ f_1^* f_5 |\alpha|^2 \left( 1 + |\alpha_1|^2 \right) \\
& + f_1^{*2} f_6 |\alpha|^2 |\alpha_2|^2 + f_1^* f_7 |\alpha|^2 |\alpha_2|^2 \\
& + f_1^* f_8 |\alpha|^2 |\alpha_3|^3 + c.c. \Big].
\end{aligned}
\tag{10}
$$

For spontaneous Raman process, $\alpha_1 = \alpha_2 = \alpha_3 = 0$, and $\alpha \neq 0$. Therefore, it is clear that $D_a$ is zero and hence the bunching and antibunching of photons are ruled out for spontaneous Raman processes. In other words, the input coherent state remains unchanged for spontaneous Raman processes. However, for stimulated Raman process (where $\alpha_1$, $\alpha_2$ and $\alpha_3$ may be nonzero) $D_a$ may be positive, negative and zero. Now, we give some numerical estimates to have some idea about the present investigation in Figure 1. The Figure 1 takes care the values of $D_a$ as the function of interaction time $t$ for various phase angles involving the complex amplitude $\alpha = |\alpha| \exp(i\vartheta)$. For $\vartheta = 0$, we observe that the photons are antibunched (Figure 1a). On the other hand, the bunching of photons are exhibited for $\vartheta = \pi/2$ (Figure 1b). It appears that the antibunching (Figure 1a) and bunching (Figure 1b) of

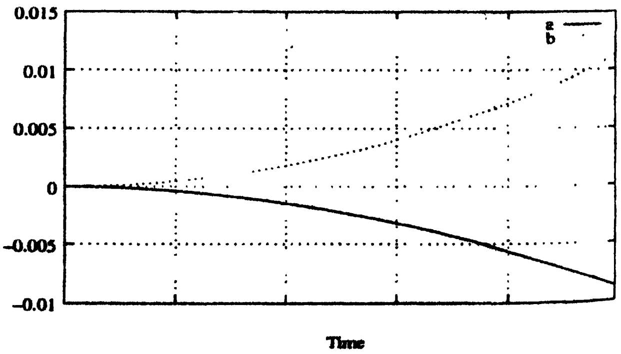


**Figure 1.** Plot of $D_a$ for the Pump mode $a$ as functions of time $t$ for $g = \chi = 0.0005$, $|\alpha| = 10$, $|\alpha_1| = 8$, $|\alpha_2| = 0.01$, $|\alpha_3| = 1$, $\omega_b = 999.999$, $\omega_c = 0.001$ and $\omega_d = 1000.0091$. For curve (a) $\vartheta = 0$ and for curve (b) $\vartheta = \pi/2$.

photons continue with the increase of time $t$. This type of monotonic nature is actually the outcome of our perturbative solutions for the field operators.

## 4. Conclusions

The quantum statistical properties of the input pump responsible for stimulated Raman scattering and for spontaneous Raman scattering are investigated. The pump mode does not exhibit the photon bunching and/or photon antibunching and hence the Poissonian photon statistics for spontaneous Raman processes. However, for stimulated Raman processes we report the photon bunching and photon antibunching. Again, the subpoissonian photon statistics is also possible for stimulated Raman processes.


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